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Thermodynamic analysis of MBE growth of quarternary InGaAsN compounds

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InGaAsN has recently been proposed as a novel material for near-infrared lasers [1]. The merits of this material are due to the strong bowing in the bandgap of the GaAs-GaN alloy system, which offers the extension of the light emission range from GaAs-based structures to 1.3 μm and longer. At the same time, the band-offsets between InGaAsN and GaAs are larger than in the conventionally used InGaAsP system, which should greatly improve high-temperature performance of 1.3 μm lasers. The combination with the presently available GaAs/AlAs distributed Bragg reflector (DBR) technology could also lead to novel vertical cavity lasers for the long wavelength region.

Up to now the best results on GaInAsN QW laser structure were demonstrated by using the MBE as a growth technique [2–5]. However, there is a wide dispersion of the published characteristics of lasers based InGaAsN quantum wells. A little attention was paid to theoretical modelling and interpretation of the growth processes of the nitride-arsenide compound. The kinetic models of growth of GaInAsN quarternary alloys were developed in Ref. [6]. However, this approach did not predict a relationship between the growth parameters and the element composition of the growing film. And the question is still actual, how incorporation of indium into GaAsN compound affects the sticking coefficient of the nitrogen.

It has been shown recently that molecular beam epitaxy can be considered in the framework of thermodynamic description under assumption that equilibrium between the gas and solid phases is established on the surface of the crystal at the substrate temperature [7, 8]. In this work we present the thermodynamic analysis of the growth of nitrogen containing ternary and quaternary Ga(In)AsN compounds. The used model allows us to predict the nitrogen mole fraction in the grown alloys as a function of external growth parameters (III- and V-group element fluxes and growth temperature).

We use the thermodynamic model developed in [9] for the analysis of the MBE growth of the quaternary (ternary) nitrogen containing compounds. The acting mass law describes the equilibrium between the gas and solid phases. The substrate temperature is taken as the system temperature. The properties of quaternary compounds are described within the model of regular solution [10]. Nitrogen atoms are assumed to be the active species.

This model predicts that dependence of the nitrogen sticking coefficient on the substrate in the case of GaAsN growth has the range of constant value which is very close to unity, i.e. complete nitrogen incorporation into the growing film (Fig. 1). In the substrate temperature range of 480–550°C strong decrease of η with increasing T was observed. The dependence of the nitrogen mole fraction on the substrate temperature demonstrates the same behaviour as the dependence of the nitrogen sticking coefficient vs substrate temperature (Fig. 2), theoretical predictions are confirmed by the experimental data with the very good precision, which is of the order of accuracy of detecting the substrate temperature. The dependence of the nitrogen fraction in the GaAsN and in the InGaAsN layers on the

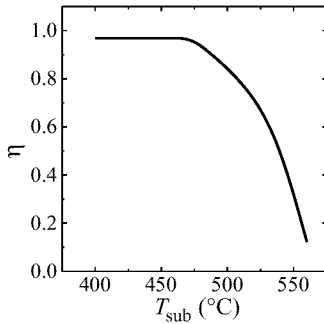


Fig. 1. The dependence of the nitrogen sticking coefficient, η , on the substrate temperature, T , for the growth of GaAsN. ($V = 1$ ML/sec, $F_{\text{As}_2}^0 = 1.5 \cdot 10^{15} \text{ cm}^{-2}\text{s}^{-1}$, $F_N^0 = 1.2 \cdot 10^{13} \text{ cm}^{-2}\text{s}^{-1}$).

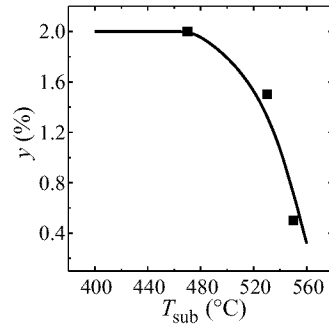


Fig. 2. The temperature dependence of the nitrogen mole fraction, y , for the growth of GaAsN. ($V = 1$ ML/sec, $F_{\text{As}_2}^0 = 1.5 \cdot 10^{15} \text{ cm}^{-2}\text{s}^{-1}$, $F_N^0 = 1.2 \cdot 10^{13} \text{ cm}^{-2}\text{s}^{-1}$).

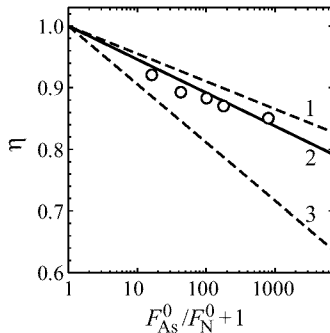


Fig. 3. The calculated sticking coefficient of the nitrogen, η , for GaAsN growth under varied flux of As and constant fluxes of N ($F_N^0 = 1.2 \cdot 10^{13} \text{ cm}^{-2}\text{s}^{-1}$) and Ga ($V = 1$ ML/s), and fixed growth temperatures (curve 1 $T = 440^\circ\text{C}$, curve 3 $T = 490^\circ\text{C}$). Experimental data for $T = 440^\circ\text{C}$ are shown as symbols and fitting curve 2.

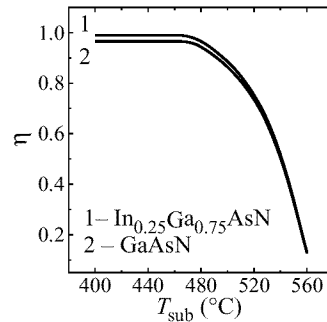


Fig. 4. The sticking coefficient of nitrogen, η , versus the growth temperature, T . ($V = 1$ ML/sec, $F_{\text{As}_2}^0 = 1.5 \cdot 10^{15} \text{ cm}^{-2}\text{s}^{-1}$, $F_N^0 = 1.2 \cdot 10^{13} \text{ cm}^{-2}\text{s}^{-1}$). Curve 1 illustrates the case of $\text{In}_{0.25}\text{Ga}_{0.75}\text{AsN}$ growth, curve 2 does the case of GaAsN growth.

growth rate was investigated (Fig. 3). In the range of high growth rates ($V > 1$ ML/s) nitrogen sticking coefficient is close to unity and nitrogen may be considered as a dopant element, i.e. $y(V)$ directly proportional to $1/V$ function. In the range of lower growth rates the nitrogen sticking coefficient becomes less than unity and $y(V)$ dependence defects from the inverse proportionality. It should be taken into account in the case of the growth of $\text{In}(\text{Ga})\text{AsN}$ quantum dots because of typically small V in this case. It was found that the nitrogen sticking coefficient depends on the ratio of the V group elements fluxes (Fig. 4) and independent of the absolute values of those parameters in the range of typical atomic nitrogen fluxes. Thus, good agreement of the theoretical predictions and the experimental data was demonstrated. Moreover, it was shown that the nitrogen sticking coefficient is determined by the total group III elements flux, i.e. adding indium (keeping the constant

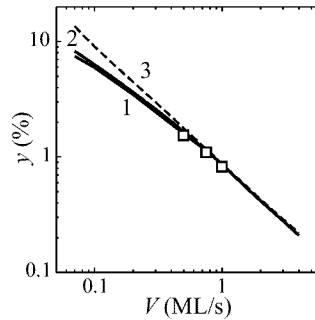


Fig. 5. The calculated dependencies of the nitrogen alloy content, y , versus growth rate, V , for the cases of growth of GaAsN (curve 1) and $\text{In}_{0.25}\text{GaAsN}$ (curve 2) with fixed $T = 450^\circ\text{C}$, $F_{\text{As}_2}^0 = 1.5 \cdot 10^{15} \text{ cm}^{-2}\text{s}^{-1}$ and $F_N^0 = 1.2 \cdot 10^{13} \text{ cm}^{-2}\text{s}^{-1}$. Curve 3 illustrates dependence which is directly proportional to the ratio of the atomic nitrogen external flux, F_N^0 , to the total III-group element external flux (which is equivalent to the V).

total growth rate) does not change significantly nitrogen incorporation into the growing film (Fig. 5).

Acknowledgements

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